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Canada Centre for Mineral and Energy Technology | Centre canadien de la technologie des minéraux et de l'énergie

CARBONIZATION RESEARCH FOR CANADIAN INDUSTRY

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by

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ABSTRACT

The Canada Centre for Mineral and Energy Technology, CANMET, is the scientific and technical branch of the Department of Energy, Mines and Resources Canada. It has the only coal carbonization research facilities in Canada with operations at the Energy Research Laboratories in Ottawa, Ontario and at the Coal Research Laboratories in Edmonton. These laboratories, which have pilot-plant and laboratory facilities for carrying out technical scale carbonization experiments and for providing a comprehensive analysis of coal properties, collaborate closely on joint programs with both the steel producers and the metallurgical coal export industry in Canada.

In addition, CANMET's carbonization scientists conduct research to develop technologies that will improve the quality of coke made from Canadian coals or Canadian steel company blends. Research topics summarized in this report include studies with Canadian coals, partial briquetting of coke oven charges and methods for predicting coke quality from coal properties.

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LA RECHERCHE SUR LA CARBONISATION POUR L'INDUSTRIE CANADIENNE

par

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RÉSUMÉ

Le Centre canadien de la technologie des minéraux et de l'énergie, CANMET, est la direction scientifique et technique du ministère de l'Énergie, des Mines et des Ressources. Au Canada, CANMET est le seul à avoir des centres de recherches sur la carbonisation du charbon en opération aux laboratoires de recherches sur l'énergie situés à Ottawa, en Ontario et aux laboratoires de recherche sur le charbon à Edmonton. Ces laboratoires, aménagés de façon à pouvoir effectuer des expériences sur la carbonisation à l'échelle pilote et de laboratoire, collaborent étroitement à des programmes conjoints avec les producteurs d'acier, de même qu'avec l'industrie d'exportation de charbon métallurgique au Canada.

De plus, les chercheurs scientifiques de la section de carbonisation du CANMET poursuivent des recherches pour mettre au point des technologies qui amélioreront la qualité du coke fait à partir de charbons canadiens ou de mélanges conçus par des compagnies canadiennes de l'acier. Les sujets de recherche résumés dans le présent rapport comprennent les études effectuées sur des charbons canadiens, le briquetage partiel de charges de fourneaux à coke et des méthodes pour prévoir la qualité du coke d'après les caractéristiques du charbon.

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INTRODUCTION

The Canada Centre for Mineral and Energy Technology (CANMET) is a scientific and technical arm of the federal Department of Energy, Mines and Resources of the Government of Canada. It serves the function of providing technical advice to government and industry on all aspects of mineral and energy technologies. It maintains a major research and development program which allows the development of internal technologies and ncecessarily emphasizes a knowledge of the current "state-of-the-art in many areas. The areas of research covered by CANMET are shown in Table 1.

CANMET houses the only centre in Canada - and one of the few on the continent - capable of fully evaluating the potential of coals for coke manufacture. The cokemaking facilities at CANMET provide technological support for the needs of the Canadian coal and steel communities. The expertise that has been built up over 60 years represents a vital contribution to technological progress in Canada's coal and steel industries.

BACKGROUND OF CANADA'S COAL AND STEEL INDUSTRIES

Canada has abundant coking coal resurces which, with the exception of a highly fluid high volatile bituminous coal from Nova Scotia, comes primarily from the two most western provinces of Canada, Alberta and British Columbia. The principal coking coal deposits lie along a 1000 km belt north of the USA-Canada border and paralleling the eastern edge of the Rocky Mountains, Figure 1. The coals are mainly low and medium volatile bituminous rank although high volatile and semi-anthracites also occur. The medium and low volatile bituminous coals generally make strong cokes when carbonized alone in spite of the petrographic high-inert contents of the coals.

In south-eastern British Columbia, the coals in the Kootenay formation are late jurassic to early cretaceous in geological age and generally have high inerts, low sulphur and moderately high ash contents. Further north, coals in the Luscar formation are lower cretaceous in age having high inerts, low sulphur contents and generally less mineral matter than the Kootenay coals.

The coking coals of western Canada are adversely located with respect to domestic and export markets; situated about 1200 km from tidewater and 4000 km from the steel mills in central Canada. Canada's bituminous coal production in 1981 grew to 22 million tonnes, the majority of which was western Canadian coking coals for export to the Pacific Rim markets (1). Overland transportation costs have been minimized by using a system of unit trains.

The integrated steel industry of Canada is located mainly in central Canada relatively near to the Appalachian coal fields of the United States and has relied historically on American sources for about 7 million tonnes of coking coals each year. Coke is generally made from blends of several high and low volatile bituminous coals. Typical properties of a Canadian steel company blend are given in Table 2. Specifications on the cokes from these blends are given in Table 3.

Coking production is the largest single cost item in the producton of steel and any improvements in its production and quality can make a big financial difference to the industry.

CARBONIZATION ACTIVITIES IN CANMET

Cokemaking research at CANMET includes evaluation of the coking properties of coals, innovations to conventional cokemaking, new methods of coke making, and related subjects. CANMET works closely with the coal and steel industries as an associate member of the Canadian Carbonization Research Association (CCRA) and provides all experimental facilities and scientific and technical support for this organization's research activities. The association has a Board of Directors and a Technical

Committee which has representatives from all major Canadian steel mills, a tar bi-products company, and most Canadian companies mining bituminous coal. CANMET scientists participate in the formulation, and planning of CCRA projects and are totally responsible for supervising the work and the reporting of results as well as being responsible for in-house carbonization projects.

The coal and carbonization facilities are housed at the Energy Research Laboratories (ERL) in Ottawa and the Coal Research Laboratories (CRL) in Edmonton, Alberta. The facilities in Ottawa provide a comprehensive description of coals used in carbonization starting from the bench scale determination of coal analysis, ash and trace element analysis, thermal rheology, and coal petrography. The laboratories at both Ottawa and Edmonton carry out technical scale carbonization experiments using 250 kg - 350 kg charges of coal in electrically or gas heated technical-scale coke ovens.

Carbonization Research

This group is responsible for the design, implementation and evaluation of both CANMET and CCRA projects. It uses the expertise and facilities provided by many other groups. Major programs in the recent past have included studies on coke quenching, partial briquetting of coke oven charges, additives to coke oven charges and coke quality from industrial and technical scale coke ovens. The group has studied the effects of ash content in western Canadian coals on coke quality and investigated methods of predicting coke quality from coal properties. It maintains records of the quality of Canada's coking coal reserves.

Conventional Cokemaking

This group is responsible for the day-to-day operation of all bench and pilot-sale ovens used in the research and quality assessment programs undertaken in Edmonton and Ottawa. The field of expertise includes coal preparation, coal sampling and physical and thermal rheological properties of coal as well as technical scale coke oven design, maintenance, operation, and coke evaluation.

This group provides an important reference source for the testing of all Canadian metallurgical coals intended for use in the Pacific Rim market.

Coal Analysis Group

This group performs all coal analyses other than rheology and petrography; the analyses relate to all CANMET research programs in combustion, gasification, liquefaction as well as carbonization. It is the major source of Canadian experts who serve on both the American Standard Testing Materials (ASTM) and the International Standards Organization (ISO) committees dealing with coal analysis. It is also the reference laboratory for the Coal Analysis Round Robin Program which involves 32 Canadian coal/analytical laboratories.

TECHNICAL SCALE COKE OVENS

A major problem with any process-oriented applied research is the choice of an appropriate scale at which to work. The scale must be sufficiently large to allow realistic extrapolation to the industrial scale; it must also be sufficiently small to allow economy and rapidity in the experimental program.

In CANMET and elsewhere this selection of scale has led to the use of carbonization ovens with a working volume of 0.25 m^3 to 0.45 m^3. At typical charge densities of 750 kg/m^3 coal capacities are 180 kg - 340 kg. The characteristic dimensions of the four CANMET ovens are presented in Table 4. The selection of this physical scale allows widths similar to those used in industrial slot-type ovens. The heat transfer characteristic between the oven wall and the coal charge can be simulated reasonably accurately. The consequent progression of thermochemical and thermophysical changes within the charge follow the stages observed in industrial ovens.

RESEARCH ACTIVITIES

Technical Scale and Industrial Oven Correlation

A continuing concern of research using technical scale ovens is the maintenance of established correlations with industrial practice. One research program has compared the product coke quality in the commercial ovens of the four Canadian steel companies with the coke produced from the same feed coal using the CANMET technical scale ovens. Although differences in coke quality are found for cokes made in different CANMET ovens, these differences are minor when the ovens are run at their standard operating conditons (3). Under standard operating condition, all technical scale ovens produce cokes with comparable strength to wharf coke from Canadian batteries. Studies of this type are currently being extended internationally in scope and an exchange of 3000 kg coal and coke samples between Canadian and Japanese steel mills has been made for round-robin testing at CANMET and Japenese research facilities. Such investigations are clearly essential to the successful extrapolation of technical-scale research data to full-scale operations and to establish technical credibility around the world.

Partial Briquetting of Coke Oven Charges

The technique of partial briquetting in cokemaking is one in which part of the charge is compressed into small briquettes and charged with the matrix coal. The technique has historically been applied to low fluidity coal blends and is in industrial use in the Japanese steel industry.

CANMET studies of partial briquetting focus on the inter-relationships between briquette concentration, charge bulk density, and coal and coke quality (4). It has been reported that the technology would not be beneficial to the relatively fluid Canadian commercial cokemaking blends. The results obtained with the commercial Canadian blends showed that partial briquetting does improve the quality of coke from Canadian commercial blends. The four-point improvement in coke stability recorded indicates a potential decrease in coke rate of about 30kg/tonne of hot metal with an associated increase in blast furnace

Other studies have shown that carbonization of partially briquetted charges at elevated flue temperature can improve coke oven through-put by 25% - 40% without any deterioration to coke quality. However, wall pressures may be excessive for some partially briquetted industrial blends carbonized at fast heating rates (5).

The results from another investigation showed that it was possible to substitute the low-volatile bituminous coal component of a fluid blend with less expensive high-volatile bituminous coal without sacrificing the coke strength (4). The use of non-coking materials as a component of the briquette alone was also investigated. The results showed that up to 70% of the briquette contents (20% of the total blend) could consist of some types of non-coking materials before coke quality deteriorated below the base level for the non-briquetted charge, Figure 2. The data developed during this program provides the technical basis on which a partial briquetting facility could be assessed for application in the Canadian steel industry.

Recent investigations (6) have shown that western Canadian coals are particularly well suited to this technology showing large improvements to ASTM and JIS coke strength indices upon partial briquetting the charges, Figure 3.

Addition of Pitch Materials to Coke Oven Charges

Several types of commercially available pitch additives have been added to a typical commercial blend used by one of central Canada's steel producers. These blends consist of high and low volatile bituminous coals from the Appalachian coal fields in the USA. Typical Canadian blend properties are listed in Table 2. Although some pitches increased thermal rheological properties more than others, no significant improvements were found when any of these pitches were added at a 5% level to the highly fludid blend (7). Apparently, blend properties are already optimized or at least require no additinal caking power.

In another study using a low fluid (high inert) western Canadian medium volatile bituminous coal, addition of 10% of experimental CANMET pitch improved coke quality substantially as shown in Table 5 (8). More recent studies have substantiated that the selection of coal for use with pitch additives is important and best improvement for medium volatile coals was found for low fluid coals of higher ranks (9).

Cokemaking with Canadian Coals

An investigation of major long-term significance was undertaken by CANMET and CCRA to determine if binary blends of commercial western and eastern Canadian coals could satisfy the quality criteria imposed by current blast furnace practice. The target quality criteria are defined in Table 3. Figures 4 and 5 show to what extent four western Canadian coals when blended with eastern Canadian coal could meet the ash, sulphur and stability criteria (10).

All blends investigated generated low-coking pressures and are expected to carbonize in conventional ovens without creating new problems. Selected blends were capable of producing cokes chemically and physically suitable for the blast furnace; in most instances it can be anticipated that a coke-rate penalty due to ash levels would be incurred.

Deviation from the optimum quality defined in Table 3 is report to have the following consequences for blast-furnace operation:

1 stability unit	8 kg/tonne hot metal and	0.8% productivity
1% ash	15 kg/tonne hot metal and	3% productivity
0.1% sulphur	4 kg/tonne hot metal and	0.8% productivity

Ash content contributes a large penalty to blast furnace coke rates and productivities.

CANMET has also investigated the effects of mineral matter on coke quality for two medium volatile, a low volatile, and a high volatile coal from western Canada (11). Each coal was beneficiated to commercially feasible ash levels and to levels about 2% higher and 2% lower than the commercial level. Thermal rheological properties were only moderatly effected by changes in mineral matter content although petrographic analysis suggested that the vitrinites were slightly more concentrated in the cleaned coal products.

The coke strength as measured by the ASTM stability factor, the prime index of coke quality in North America, is remarkably dependent on the ash content, particularly so for the medium volatile coals as shown in Figure 6. When each of these coals was 50% blended with a good coking coal blend the rates of change with each percent of ash decreased slightly. Other changes to coke properties caused by reduced ash include; higher coke hardness; smaller mean size and less reactivity to carbon dioxide. Removal of large shale particles appeared responsible for substantially increased strength of coke made from at least one of the medium volatile coals (Coal H). Coke microscopy showed that ash reduction did not affect substantially the types of optical anisotropy of the coke.

Prediction of Coke Quality from Western Canadian Coal Properties

Several methods have originated in different areas of the world for predicting coke strength from coal properties. CANMET scientists have examined the applicability of several of these methods to western Canadian coals (12). Methods examined include: petrography (13); G Factor Method (14); coke strength versus volatile matter content and dilatation (15, 16). Data from western Canadian coal and coke properties have also been regressed in attempts to develop better models for these coals.

Petrographic analysis appears to be the best of the established methods for predicitng coke quality from coal properties for western Canadian coals. Methods based on dilatation properties cannot be used for many western Canadian coals which have poor dilatation characteristics but produce strong cokes. By using regression analyses based on coal properties and technical scale oven results from a decade of work, CANMET scientists have developed other methods for predicting coke strengths for western Canadian coals. One of the better methods illustrated in Figure 7, uses fludity and reflectance (modified MOF diagram) to predict coke strength. The figure shows that for low rank coals R_o <1.0) stability is strongly rank dependent, but for higher rank coals fluidity becomes predominant. This method appears to work with blends of Canadian coals as well as for pure coals.

Prediction of Coke Characteristics for Blends of Chinese and Canadian Coals

CANMET scientists have carried out paper exercises to predict the quality of coke that could be expected in various blends of indigenous and foreign coking coals. The objectives of the calculations were to define the approximate coke stability factor, ash content and sulphur levels for various coal blends. Predictive methods include:

1. Inerts/Reflectance Method - which uses maceral compostion analysis and mean vitrinite reflectance to define coke stability as shown in Figure 8 (12, 17).

2. Composition Balance/Strength Index Method - which uses petrographic reflectance distribution of the vitrinite macerals and maceral composition analysis to calculate petrographic composition balance and strength indices to define coke stability as shown in Figure 9.

3. Fluidity/Reflectance Method - which has been developed by CANMET scientists for use with western Canadian coals as shown in Figure 7 (12).

Examples of coke quality predictions for six Canadian and three Chinese coals and for various binary blends of these coals are given in Table 7 and Figures 8, 9 and 10. It should be remembered that predictions are based on limited literature data on the Chinese component coals of the blends. Table 8 presents the coal analyses on which the predictions were based.

The predictive data developed here indicates that many Chinese-Canadian coal blends can meet the stability and sulphur standards required by current Canadian blast furnace practise, Table 3. Ash contents of the coke may be marginally above desired levels.

Analytical and predictive data could only be substantiated by co-operative experimental investigation utilizing technical scale coke ovens.

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Table 1 - CANMET energy research and development program

ENERGY RESEARCH PROGRAM

Resource Assessment	-	Oil and Gas
	-	Coal and Peat
Conservation Technology	-	Transportation
	-	Oil and Gas Combustion
Petroleum Supply	-	Bitumen/Heavy Oil Recovery
	-	Bitumen/Heavy Oil Upgrading
	-	Catalyst Development
	-	Bitumen/Heavy Oil & Product Characterization
Coal Technology	-	Coal Preparation
	-	Carbonization
	-	Gasificaiton
	-	Liquefaction
	-	Combustion
	-	Coal Slurry Technology
	-	Coal Analysis
Renewable Energy Sources	-	Combustion
	-	Conversion

Table 2 - Typical analyses of a Canadian steel industry blend

Coal Blend Properties	Canadian
Chemical Analysis, (db) %	
Volatile Matter	27 - 30
Ash	4 - 7.5
Fixed Carbon	63 - 68
Gieseler Maximum Fluidity, ddpm	700 - 2500
Mean Maximum Reflectance, Ro, %	1.07 - 1.17

Table 3 - Typical specificaitons for Canadian blast furnace coke

Stability Factor	57
Ash Content	7%
Sulphur Content	0.6% or less
Phosporous Content	0.1% or less
Alkali	0.1% or less
Apparent Specific Gravity	0.89 - 0.91

Table 4 - Characteristics of the CANMET technical scale ovens

Oven Designation:	310 mm Oven	460 mm Oven (rebuilt 1976)	Koppers	Carbolite
Design Basis	Associates	CANMET	Koppers	Carbolite
Location	Ottawa	Ottawa	Edmonton	Edmonton
Date Installed	1971	1976	1973	1979
Operating 1983	yes	yes	yes	yes
Oven Construction:				
Movable Wall	yes	yes	yes	yes
Coking Chamber width; cm	30.5	30.5	30.5	Variable; 30-47
Coke Chamber Refractories	Silicon Carbide	High Density Silica	Alcor	Silicon Carbide
Heating Method	Glow Bars (Electric)	Glow Bars (Electric	Natural Gas	Glow Bars (Electric)
Standard Oven Test Conditions:				
Charge Weight, kg	250	350	200	350
Charge Pulverization; % -6 mesh	80 ± 5	80 ± 5	80 ± 5	80 ± 5
Target Charge Moisture; %	2.0	6.0	2.0	6.0
Dry Bulk Density, kg/m^3	816 ± 16	744 ± 16	816 ± 16	800 ± 16
Flue Temperature Control	900°C to 1070°C @ 32°C/min	constant 1125°C	constant 1077°C	875 to 1130 @ 15°C/min
Charge Push Method*	0.5 hr after C_T = 1010°C	0.5 hr after C_T = 1000°C	0.5 hr after C_T = 1010°C	3.0 hr after C_T = 950°C
Normal Push Time; hr	9	18	18	18

*C_T = Charge Centre Temperature

Table 5 - Comparison of technical scale carbonization test results for MVb coal and MVb coal with 10% pitch

	MV Coal	MV Coal + 10% CANMET Pitch
Test Rate		
Moisture of Charge, %	3.0	2.4
% Minus 3 mm	89.1	90.2
Oven Bd (db) kg/m^3	805	808
Gross Coking Time h:min	9:05	9:45
Coke Yield, %	75.2	75.0
Mean Coke Size mm	48.0	51.3
Apparent Spec. Gravity	0.96	0.98
Breeze (-12 mm), %	3.6	3.1
ASTM Stability Factor	45.5	56.8
Hardness Factor	63.9	69.5
JIS DI_{15}^{30}	91.8	94.4
DI_{15}^{150}	77.5	82.3

Table 6 - Rank and proximate analysis of cleaned coals

	Coal G			Coal H			Coal I			Coal J		
Ash	High	Med	Low	High	Med	Low	High	Med	Low	High	Med	Low
ASTM Rank	mvb	mvb	mvb	mvb	mvb	mvb	hvAb	hvAb	hvAb	lvb	lvb	lvb
International System	431	431	432	432	433	433	633	634	634	421	432	433
Carbon (dmmfb)	89.1	88.8	88.8	91.7	90.8	90.9	86.8	86.5	86.2	92.0	91.4	90.9
Proximate Analysis (db)												
Ash %	8.2	7.1	5.3	7.9	5.7	3.1	9.6	6.6	5.1	11.2	8.9	6.8
Volatile Matter	24.8	25.1	25.8	22.9	23.5	23.8	30.6	32.2	30.2	19.6	20.4	21.2
Fixed Carbon	67.0	67.8	68.9	69.2	70.8	73.1	59.8	61.2	64.7	69.2	70.7	72.0

Table 7 - Predicted coke quality characteristics: Binary Blends

Blend Composition	Predicted ASTM Stability Method 1	Method 2	Method 3	Predicted Coke Ash Content, %	Sulphur Content, %
100% Zao Zhuang					
70% Zao Zhuang:30% K	60	52-56	56	11.1	0.60
70% Zao Zhuang:30% L	56	48-52	46	11.5	0.60
50% Zao Zhuang:50% L	49	50-55	52	11.3	0.55
70% Zao Zhuang:30% M	45	28-32	43	12.1	0.58
50% Zao Zhuang:50% N	55	52-54	54	12.1	0.50
100% Kai Luan	60	62	55	15.2	0.70
70% Kai Luan:30% K	64	58-60	60	13.4	0.60
70% Kai Luan:30% L	60	60	57	13.8	0.60
70% Kai Luan:30% M	57	55-57	55	14.4	0.58
50% Kai Luan:50% M	55	52-56	55	13.8	0.50
50% Kai Luan:50% N	60	57-59	59	13.7	0.51
70% Kai Luan:30% Yan Zhou	56	53-58	54	14.0	0.60
100% Yan Zhou	5	5	30	10.5	0.36
70% Yan Zhou:30% K	50	47-52	42	10.2	0.38
50% Yan Zhou:50% N	50	43-47	48	11.5	0.34
70% Zao Zhuang:30% O	56	52-56	53	10.8	0.60
70% Kai Luan:30% O	60	53-57	59	13.1	0.60
70% Yan Zhou:30% O	47	45	43	9.8	0.36
50% Zao Zhuang:50% P	59	61	53	9.5	0.60
50% Kai Luan:50% P	64	59	59	11.2	0.60
50% Yan Zhou:50% P	53	50	49	8.7	0.43

Table 8 - Analysis of individual blend components

	Proximate Analysis, %				Coking Characteristics				
	A (db)	FC (db)	S (db)	VM (db)	F.S.I.	Gieseler Fluidity ddpm	R_o	Contraction/ Expansion	Total Dilatation
Chinese Coking Coals									
Zao - Zhuang	8.2	57.87	0.69	38.9	5½/7	6-13,000	0.92	$\frac{25-30}{42-77}$	72-102
Kai - Luan	11.7	63.77	0.70	24.6	6½/8	4,450	1.30	$\frac{26-31}{100-170}$	130-196
Yan - Zhou	7.2	58.27	0.45	34.5	2½	35	0.76	-	-
Canadian Coking Coals									
K	8.0	73.4	0.44	18.6	4	2.5	1.58	14/11	25
L	8.4	68.1	0.39	23.5	7½	79	1.27	24/16	40
M	9.5	65.0	0.3	25.5	5	435	1.06	23/49	72
N	9.8	68.4	0.32	21.8	7	20	1.38	16/23	39
O (Developing Mine)	7.0	72.5	0.36	20.0	2½	1	1.60	-	0
P (Developing Mine)	5.6	69.7	0.49	24.7	7½	138	1.38	18/30	48

A = ash, FC = FC, S = Sulphur, VM = Volatile Matter

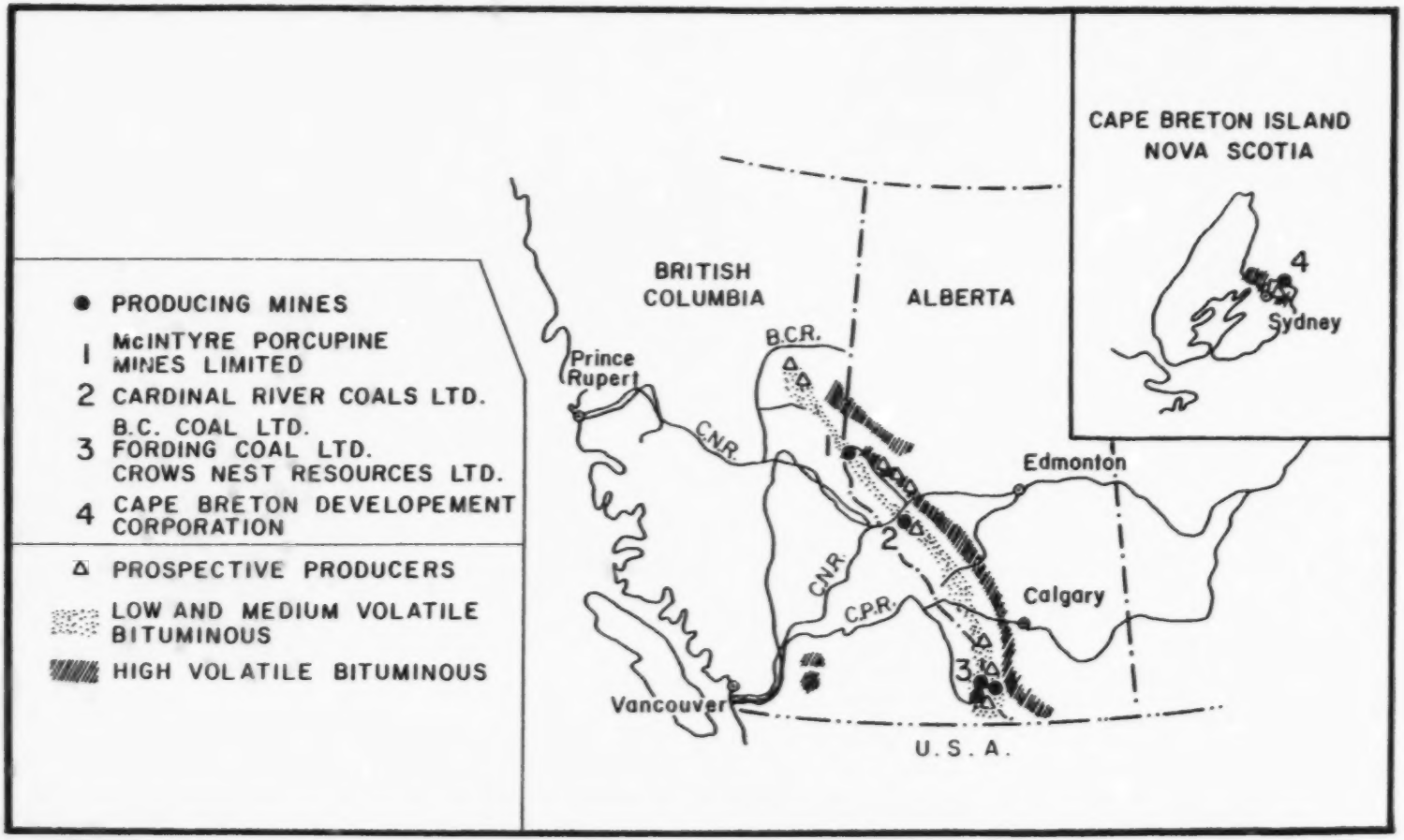

Fig. 1 - The general distribution of coking coals in western Canada and the location of principal coking coal mines

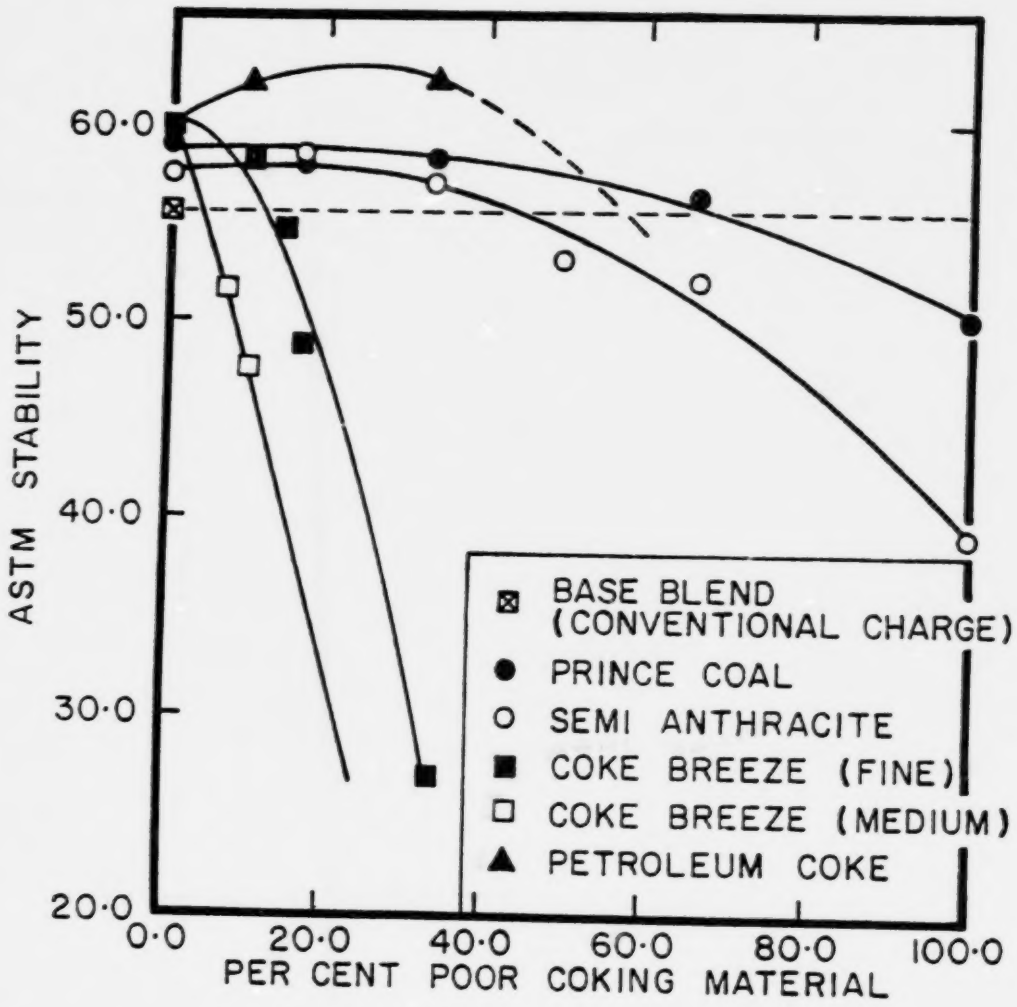

Fig. 2 - The use of non-coking materials in briquette additives to partially briquetted coal charges

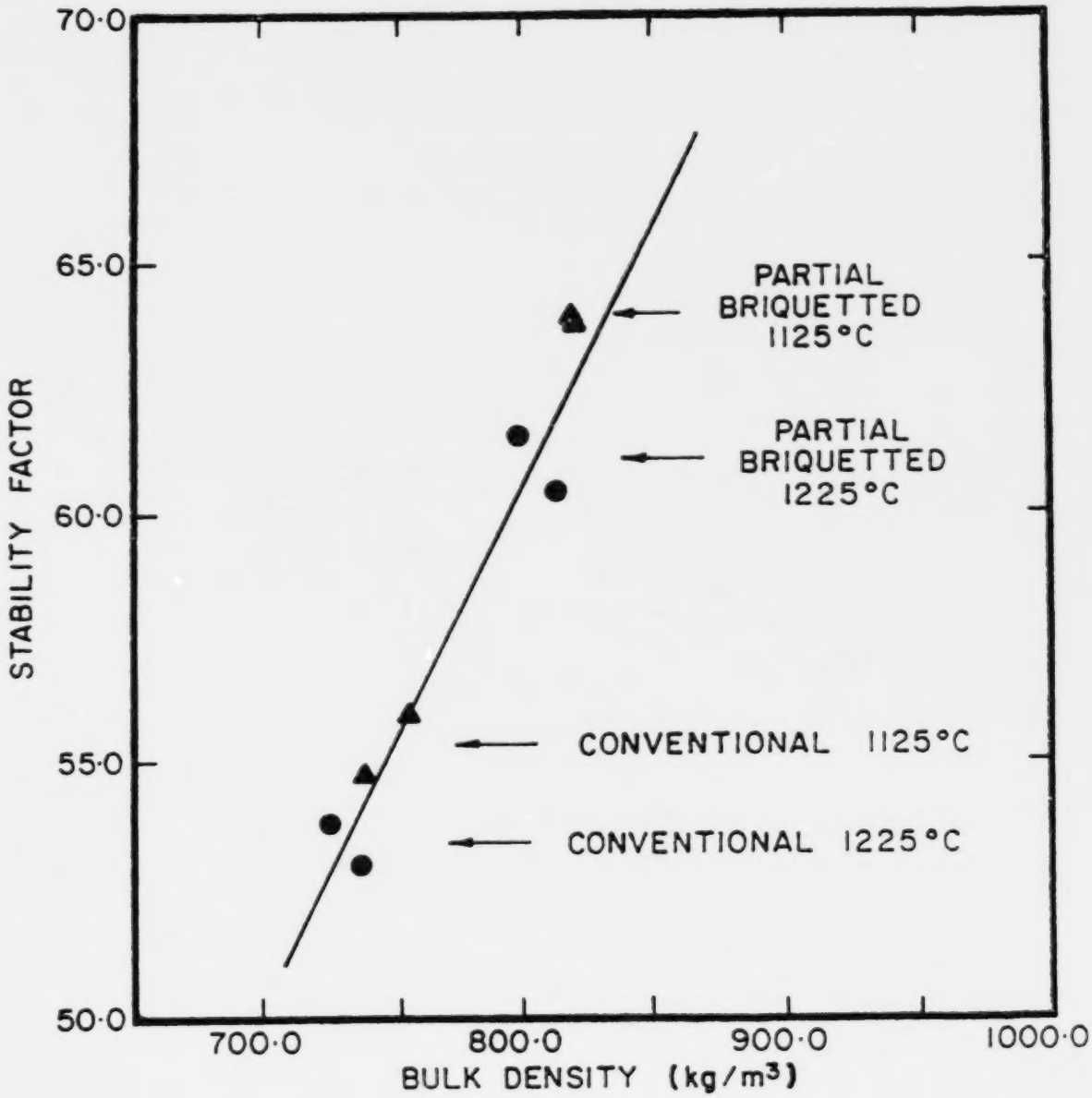

Fig. 3. Effect of changes in the coal bulk density in the 460-mm coke oven on the coke ASTM stability factor for coke made from western Canadian medium volatile coal by conventional and partially briquetted charges.

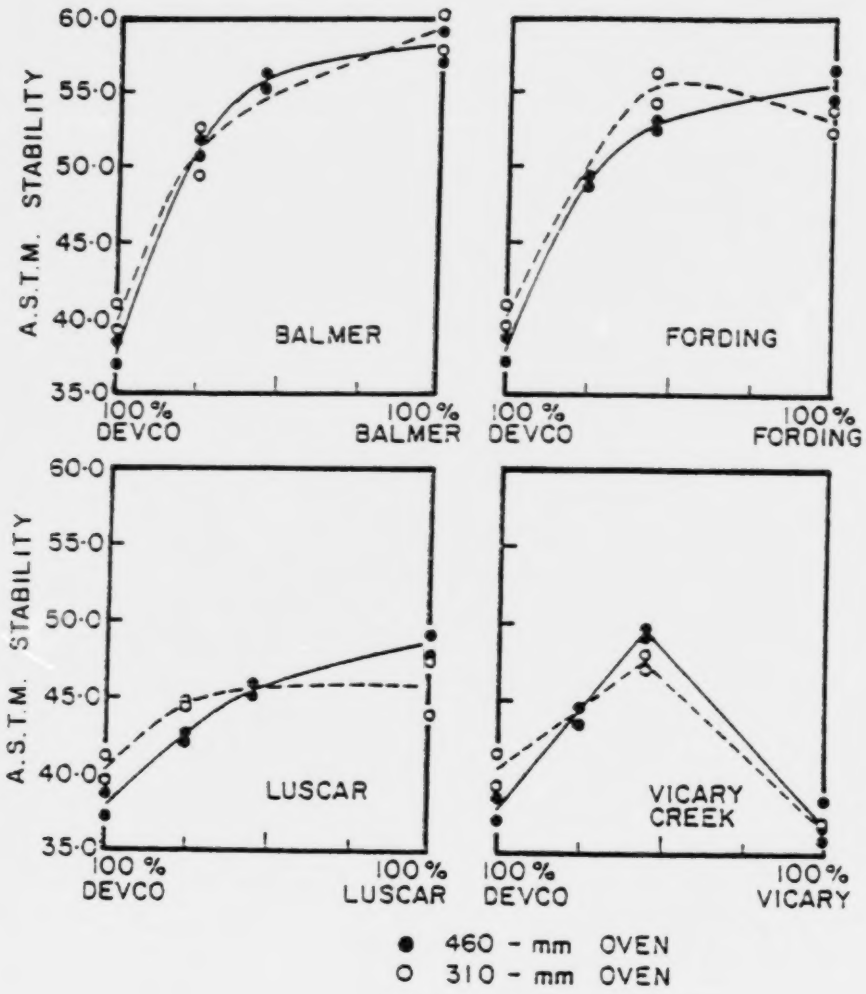

Fig. 4 - Relationship between stability factor and blend composition

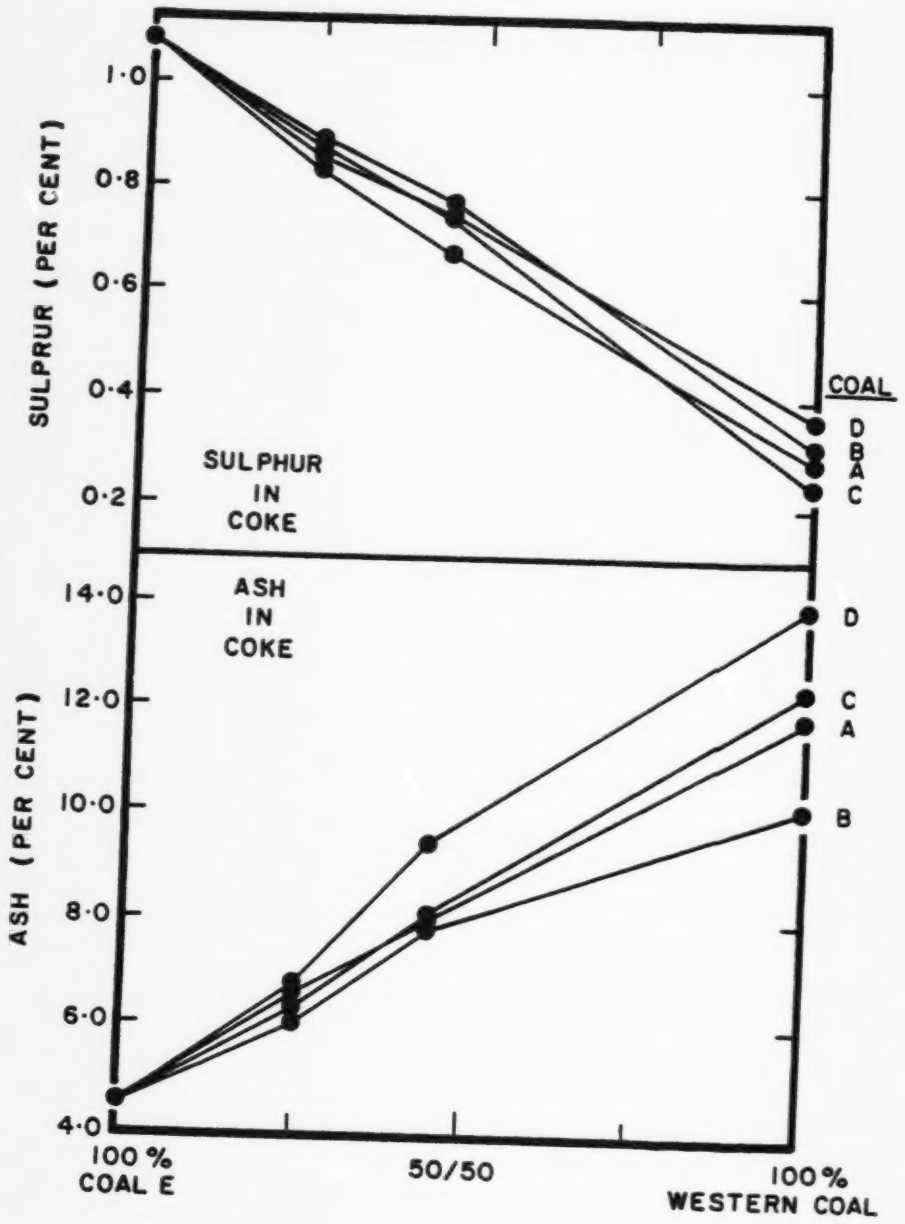

Fig. 5 - The relationship between stability factor and blend composition for binary Canadian coal blends

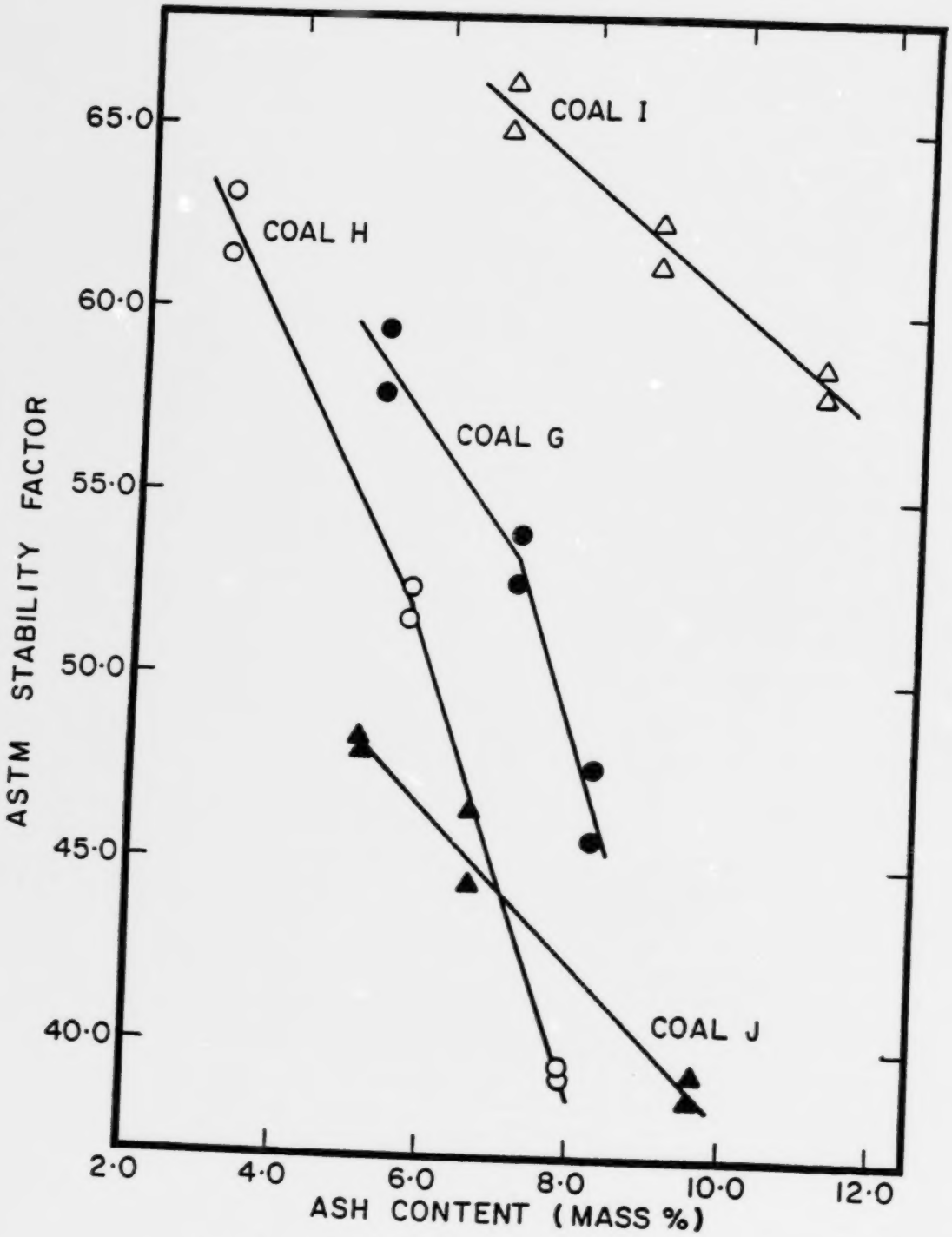

Fig. 6 - ASTM coke stability factor versus ash content of coal - Carbolite oven

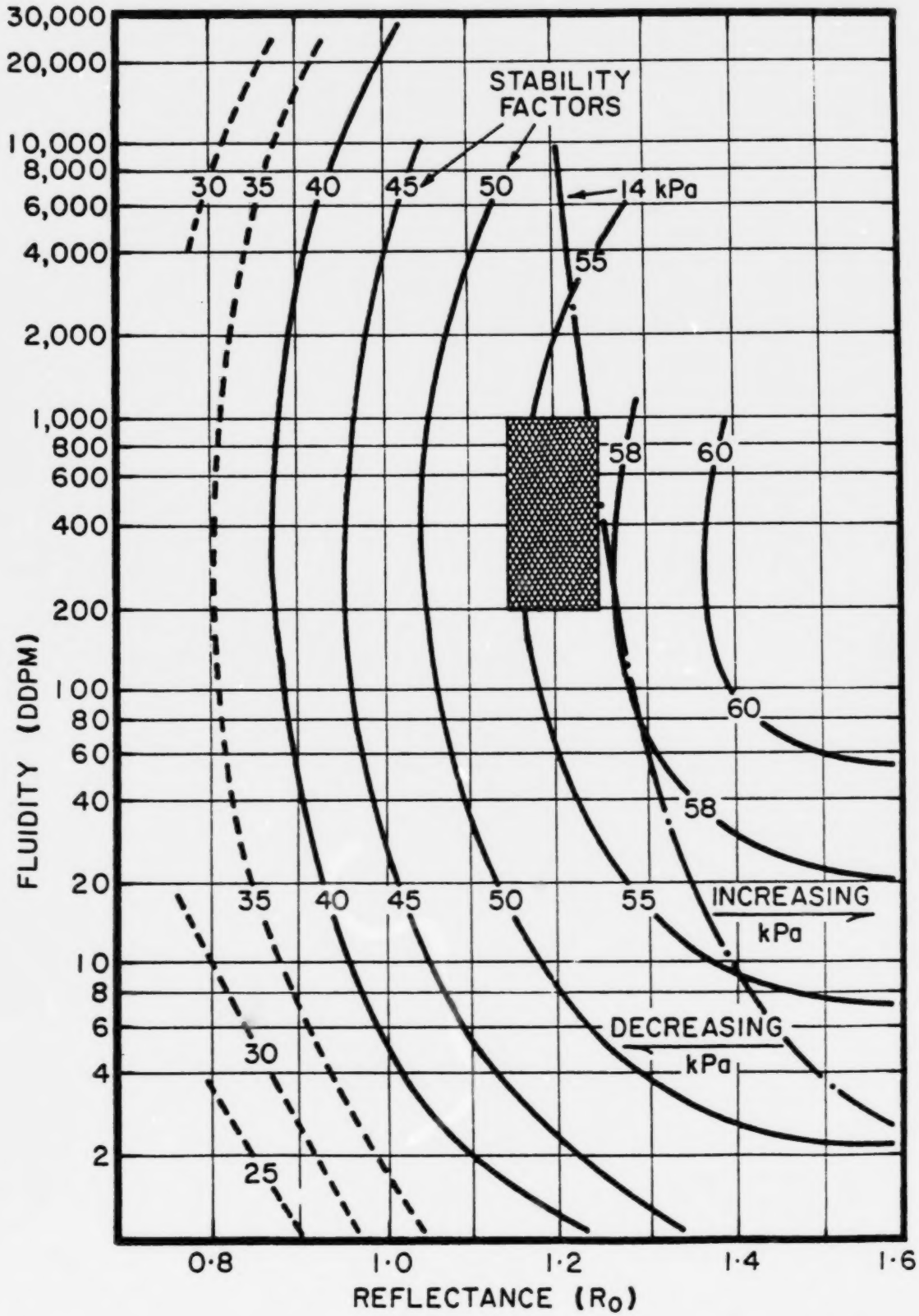

Fig. 7 - The relationship between Gieseler fluidity, mean-maximum vitrinite reflectance and coke stability for western Canadian coals

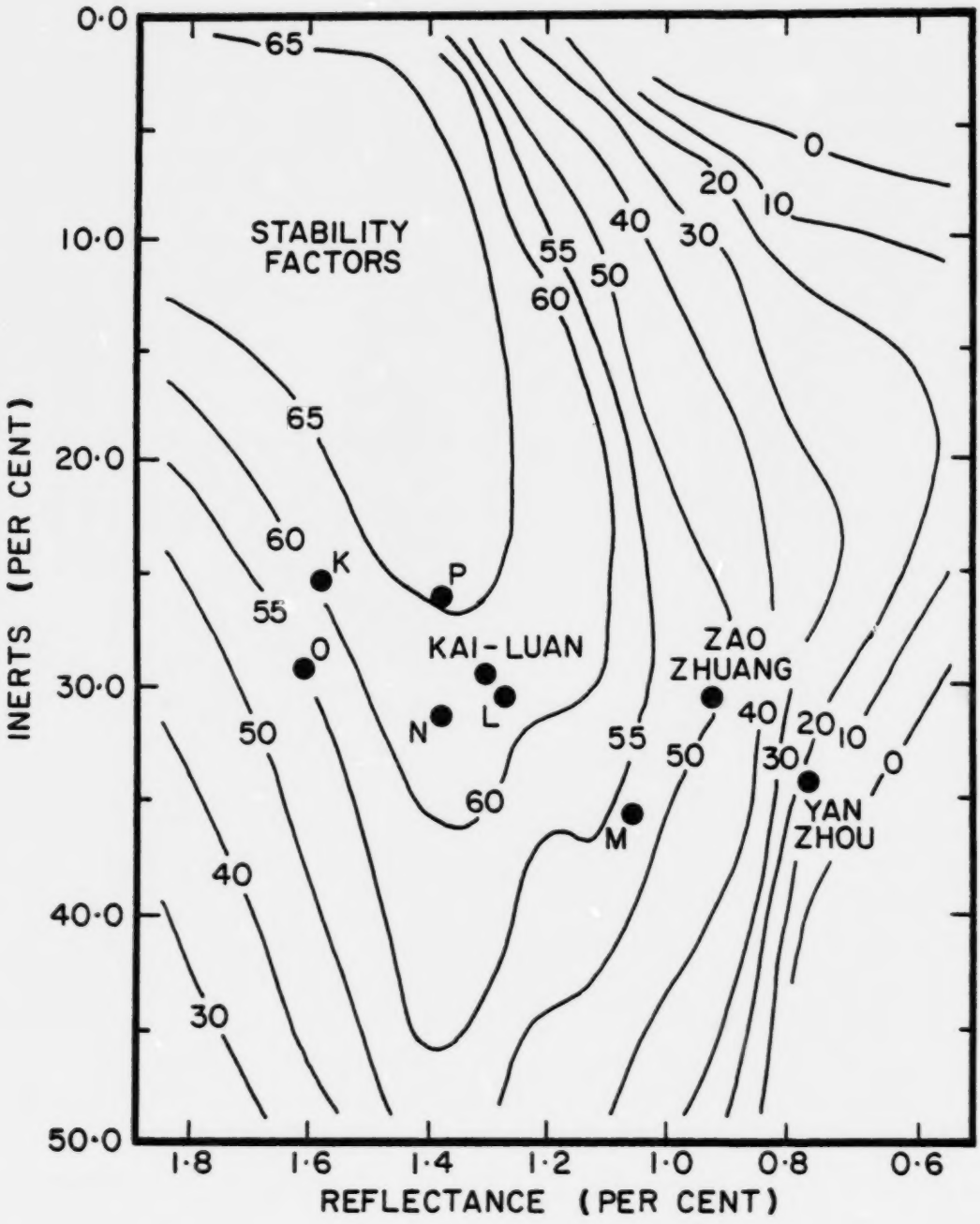

Fig. 8 - Relationship between coke stability factor with coal reflectance and petrographic inerts

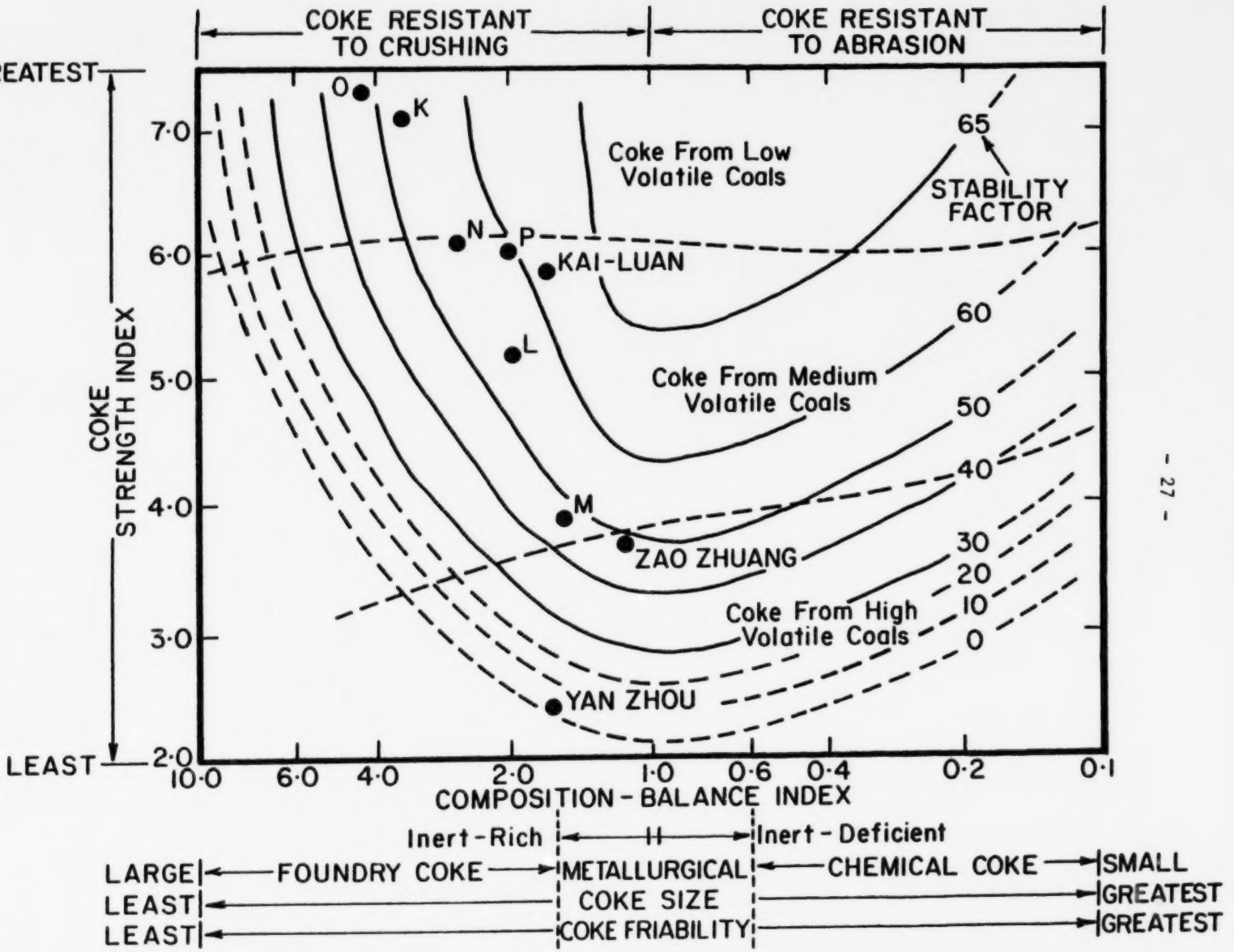

Fig. 9 - Petrographic method of predicting coke strength. Position of Chinese coals are estimates

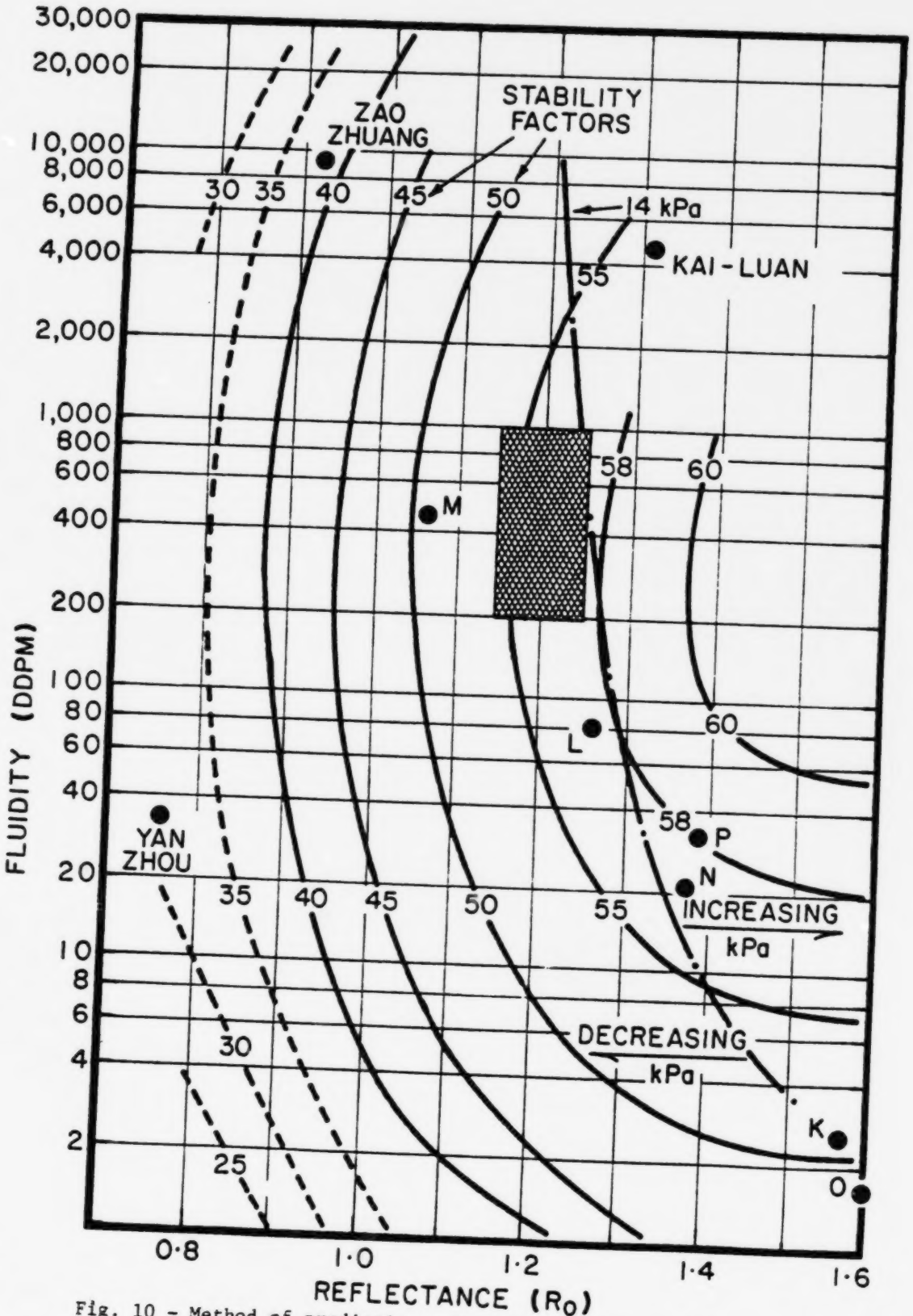

Fig. 10 - Method of predicting coke strength from coal fluidity and mean maximum reflectance - based on CANMET regression analyses for western Canadian coals